

# Phases of triangular lattice antiferromagnet near saturation

Oleg A. Starykh,<sup>1</sup> Wen Jin,<sup>1</sup> and Andrey V. Chubukov<sup>2</sup>

<sup>1</sup>Department of Physics and Astronomy, University of Utah, Salt Lake City, UT 84112

<sup>2</sup>Department of Physics, University of Wisconsin, Madison, WI 53706

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We consider 2D Heisenberg antiferromagnets on a triangular lattice with spatially anisotropic interactions in a high magnetic field close to the saturation. We show that this system possess rich phase diagram in field/anisotropy plane due to competition between classical and quantum orders: an incommensurate non-coplanar spiral state, which is favored classically, and a commensurate co-planar state, which is stabilized by quantum fluctuations. We show that the transformation between these two states is highly non-trivial and involves two intermediate phases – the phase with co-planar incommensurate spin order and the one with non-coplanar double- $\mathbf{Q}$  spiral order. The transition between the two co-planar states is of commensurate-incommensurate type, not accompanied by softening of spin-wave excitations. We show that a different sequence of transitions holds in triangular antiferromagnets with exchange anisotropy, such as  $\text{Ba}_3\text{CoSb}_2\text{O}_9$ .

**Introduction.** The field of frustrated quantum magnetism witnessed a remarkable revival of interest in the last few years due to rapid progress in synthesis of new materials and in understanding previously unknown states of matter. The two main lines of research in the field are searches for spin-liquid phases and for new ordered phases with highly non-trivial spin structures [1]. For the latter, the most promising system is a 2D Heisenberg antiferromagnet on a triangular lattice in a finite magnetic field, as this system is known to possess an “accidental” classical degeneracy: every classical spin configuration with a triad of neighboring spins satisfying  $\mathbf{S}_r + \mathbf{S}_{r+\delta_1} + \mathbf{S}_{r+\delta_2} = \mathbf{h}/(3J)$ , where  $J$  is the exchange interaction, belongs to the ground state manifold.

An infinite degeneracy, however, holds only for an ideal Heisenberg system with isotropic nearest-neighbor interaction. Real systems have either spatial anisotropy of exchange interactions, as in  $\text{Cs}_2\text{CuCl}_4$  [2, 3] and  $\text{Cs}_2\text{CuBr}_4$  [4–6] for which the interaction  $J$  on horizontal bonds is larger than  $J'$  on diagonal bonds (see insert in Fig. 1), or exchange anisotropy in spin space, as in  $\text{Ba}_3\text{CoSb}_2\text{O}_9$ , for which  $J_z < J_\perp = J$  (an easy plane anisotropy) [7–9]. An anisotropy of either type breaks accidental degeneracy already at a classical level and for fields  $\mathbf{h} = h\hat{z}$  slightly below the saturation field  $h_{\text{sat}}$  selects a non-coplanar cone state with

$$\langle \mathbf{S}_r \rangle = (S - \rho)\hat{z} + \sqrt{2S\rho}(\cos[\mathbf{Q} \cdot \mathbf{r} + \varphi]\hat{x} + \sin[\mathbf{Q} \cdot \mathbf{r} + \varphi]\hat{y}), \quad (1)$$

where  $\rho \sim S(h_{\text{sat}} - h)/h_{\text{sat}}$  is the density of magnons (the *condensate fraction*) which determines the magnetization  $M = S - \rho$ ,  $\varphi \in (0, 2\pi)$  is a phase of a condensate, and  $\mathbf{Q} = (Q, 0)$  is the ordering wave vector. It is incommensurate with  $Q = Q_i = 2\cos^{-1}(-J'/2J)$  in the spatially anisotropic case  $J' \neq J$  and commensurate with  $Q = Q_0 = 4\pi/3$  for the easy-plane anisotropy (in the last case, the values of  $\mathbf{Q}_0 \cdot \mathbf{r} = 2\pi\nu/3 \pmod{2\pi}$ , with  $\nu = \pm 1, 0$ ).

Quantum fluctuations are also known to lift accidental degeneracy, and do so already in the *isotropic* system. However, they select different ordered state, which is the co-planar, commensurate state with two parallel spins in every triad, often called the V state (Fig. 1) [1, 10, 11].

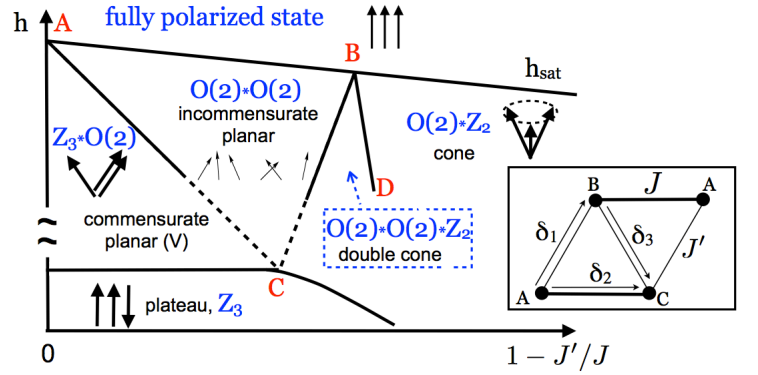


FIG. 1. Phase diagram of the spatially anisotropic triangular lattice antiferromagnet with large  $S$  near saturation field, as a function of spatial anisotropy of the interactions. The phases at small and large anisotropy are commensurate co-planar V-phase, which breaks  $Z_3 \times O(2)$  symmetry, and incommensurate non-coplanar chiral cone phase, which breaks  $Z_2 \times O(2)$  symmetry. In between, there are two incommensurate phases: a co-planar phase, which breaks  $O(2) \times O(2)$  symmetry, and a non-coplanar double cone phase, which breaks  $Z_2 \times O(2) \times O(2)$  symmetry. Line AC denotes the CI transition from the V phase to the incommensurate planar phase. The insert shows the geometry of the lattice exchange constant is  $J$  on horizontal bonds (bold) and  $J'$  on diagonal bonds (thin).

This order is described by

$$\langle \mathbf{S}_r \rangle = (S - 2\rho \cos^2[\mathbf{Q} \cdot \mathbf{r} + \theta])\hat{z} + \sqrt{4S\rho} \cos[\mathbf{Q} \cdot \mathbf{r} + \theta] \times (\cos \varphi \hat{x} + \sin \varphi \hat{y}), \quad (2)$$

where  $\mathbf{Q} = \mathbf{Q}_0$ ,  $\rho = \rho_{\mathbf{Q}_0} + \rho_{-\mathbf{Q}_0}$  is the sum of two equal contributions from condensates with wave vectors  $\pm \mathbf{Q}_0 = (\pm Q_0, 0)$ ,  $\varphi$  is a common phase of the two condensates, and  $\theta$  is their *relative* phase. The values of  $\theta$  in the commensurate V phase are constrained to  $\theta = \pi\ell/3$ , where  $\ell = 0, 1, 2$  describe three distinct degenerate spin configurations (three choices to select two parallel spins in any triad, see Fig. 1).

The issue we consider in this paper is how the system evolves at  $h \leq h_{\text{sat}}$  from the co-planar V state, selected

by quantum fluctuations, to the non-coplanar cone state, selected by classical fluctuations, as the anisotropy increases. We show that this evolution is highly non-trivial and involves commensurate-incommensurate transition (CIT) and, in the case of  $J - J'$  model, an intermediate double cone phase.

**The phase diagrams.** To begin, it is instructive to compare order parameter manifolds in the two phases. The order parameter manifold in the V phase is  $O(2) \times Z_3$  and that in the cone phase is  $O(2) \times Z_2$ . In both phases, a continuous  $O(2)$  reflects a choice of the phase  $\varphi$ .  $Z_3$  in the V phase corresponds to choosing one of three values of  $\theta$  in (2), and  $Z_2$  in the cone phase is a chiral symmetry between left- and right-handed spiral orders (chiralities), i.e. orders with  $+Q$  and  $-Q$  in (1). The symmetry breaking patterns in the two phases are not compatible, hence one should expect either first-order transition(s) or an intermediate phase(s). We show that in  $J - J'$  model the evolution occurs via two intermediate phases, see Fig. 1. As  $\delta J = J - J'$  increases, the V phase first undergoes a CIT at  $\delta J_{c1} \sim (J/\sqrt{S})(h_{\text{sat}} - h)/h_{\text{sat}}$  (line AC in Fig. 1). The new phase remains co-planar, like in (2), but the phase  $\theta$  becomes incommensurate and coordinate-dependent, and order parameter manifold extends to  $O(2) \times O(2)$  (spontaneous selection of  $\varphi$  and the origin of coordinates). The incommensurate co-planar state exists up to a second critical  $\delta J_{c2} \sim J/\sqrt{S}$ , where the system breaks the  $Z_2$  symmetry between the two condensates (line BC in Fig. 1). At larger  $\delta J$  the two condensates still develop, one of them shifts to a new wave vector  $\bar{\mathbf{Q}}$  and its magnitude gets smaller. The resulting state is a non-coplanar double cone state with order parameter manifold  $O(2) \times O(2) \times Z_2$ . Finally, at the third critical anisotropy  $\delta J_{c3} = \delta J_{c2}[1 + O((h_{\text{sat}} - h)/h_{\text{sat}})]$  the magnitude of the condensate at  $\bar{\mathbf{Q}}$  vanishes and the double cone transforms into a single cone (line BD in Fig. 1).

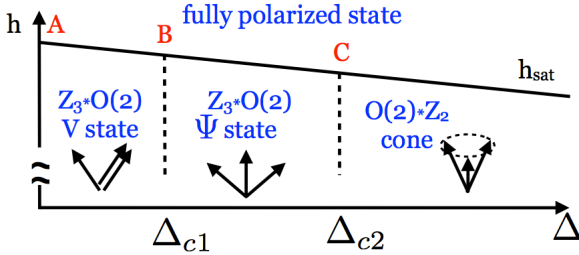


FIG. 2. The phase diagram of the XXZ model in a magnetic field near a saturation value,  $\Delta = (J - J_z)/J$ . The cone and V states are the same as in Fig. 1, but the transformation from one phase to the other with increasing spin exchange anisotropy proceeds differently from the case of spatial exchange anisotropy and involves one intermediate co-planar commensurate phase with  $\Psi$ -like spin pattern.

In systems with easy-plane anisotropy  $\Delta = (J - J_z)/J > 0$ , the ordering wave vector remains commensurate,  $Q = Q_0 = \pm 4\pi/3$ , for all  $\Delta > 0$ , and the evolution from quantum-preferred V state to classically-preferred cone state proceeds differently, via two first-order phase transitions (see Fig. 2). The V state with  $\theta = \ell\pi/3$  survives up to some critical  $\Delta_{c1} \sim$

$1/S$ , where another commensurate co-planar order develops, for which  $\theta = (2\ell + 1)\pi/6$ . The corresponding spin pattern resembles Greek letter  $\Psi$  and we label this state a  $\Psi$  phase. The  $\Psi$  phase survives up to  $\Delta_{c2} \geq \Delta_{c1}$ , beyond which the spin configuration turns into the commensurate cone state.

We now discuss the model and the calculations which lead to phase diagrams in Figs. 1 and 2.

**The model.** The isotropic Heisenberg antiferromagnet on a triangular lattice is described by the Hamiltonian

$$\mathcal{H}_0 = \frac{1}{2}J \sum_{\mathbf{r}, \delta} \mathbf{S}_{\mathbf{r}} \cdot \mathbf{S}_{\mathbf{r}+\delta} - \sum_{\mathbf{r}} h S_{\mathbf{r}}^z, \quad (3)$$

where  $\delta$  are nearest-neighbor vectors of the triangular lattice. The two perturbations we consider are

$$\delta\mathcal{H}_{\text{anis}} = (J' - J) \sum_{\mathbf{r}} \mathbf{S}_{\mathbf{r}} \cdot (\mathbf{S}_{\mathbf{r}+\delta_1} + \mathbf{S}_{\mathbf{r}+\delta_3}), \quad (4)$$

$$\delta\mathcal{H}_{\text{xxz}} = \frac{1}{2}(J_z - J) \sum_{\mathbf{r}, \pm\delta_{1,2,3}} S_{\mathbf{r}}^z S_{\mathbf{r}+\delta}^z. \quad (5)$$

where  $\langle \mathbf{r}, \mathbf{r} + \delta_{1,3} \rangle$  are diagonal bonds.

We consider a quasi-classical limit  $S \gg 1$ , when quantum fluctuations are small in  $1/S$  and quantum and classical tendencies compete at small anisotropy  $\delta J/J \sim 1/\sqrt{S}$  and/or  $\Delta/J \sim 1/S$ . In this limit, the calculations in the vicinity of the saturation field can be done using a well-established dilute Bose gas expansion and are controlled by simultaneous smallness of  $1/S$  and of  $(h_{\text{sat}} - h)/h_{\text{sat}}$  [11, 13–15]. We argue that our results are applicable for all values of  $S$ , down to  $S = 1/2$ , because (i) quantum selection of the V state holds even for  $S = 1/2$  [14], and (ii) numerical analysis of  $S = 1/2$  systems [14, 17] identified the same phases near saturation field as found here.

We set quantization axis along the field direction and express spin operators  $\mathbf{S}_{\mathbf{r}}$  in terms of Holstein-Primakoff bosons  $a, a^\dagger$  as  $S_{\mathbf{r}}^- = [2S - a_{\mathbf{r}}^\dagger a_{\mathbf{r}}]^{1/2} a_{\mathbf{r}}^\dagger$ ,  $S_{\mathbf{r}}^z = S - a_{\mathbf{r}}^\dagger a_{\mathbf{r}}$ . Substituting this transformation into  $\mathcal{H}_{\text{anis/xxz}}$  and expanding the square root one obtains the spin-wave Hamiltonian  $\mathcal{H} = \mathcal{E}_{\text{cl}} + \sum_{j=2}^{\infty} \mathcal{H}^{(j)}$ , where  $\mathcal{E}_{\text{cl}}$  stands for the classical ground state energy, and  $\mathcal{H}^{(j)}$  are of  $j$ -th order in operators  $a, a^\dagger$ . For our purposes, terms up to  $j = 6$  have to be retained in the expansion (see the Supplement [18] for technical details). The quadratic part of the spin-wave Hamiltonian reads

$$\mathcal{H}^{(2)} = \sum_{\mathbf{k}} (\omega_{\mathbf{k}} - \mu) a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \quad (6)$$

where  $\omega_{\mathbf{k}} = S(J_{\mathbf{k}} - J_{\mathbf{Q}})$  is the spin-wave dispersion, measured relative to its minimum at the saturation field  $h_{\text{sat}}$ , and  $\mu = (h_{\text{sat}} - h)/h_{\text{sat}}$  plays the role of chemical potential. For  $J - J'$  model,  $J_{\mathbf{k}} = \sum_{\pm\delta_j} J_{\delta_j} (e^{i\mathbf{k} \cdot \delta_j} - 1)$ , where  $J_{\delta_{1,3}} = J'$  and  $J_{\delta_2} = J$ . Here  $\mathbf{Q} = \mathbf{Q}_i = (Q_i, 0)$  with  $Q_i = 2\cos^{-1}(-J'/2J)$ . For XXZ model,  $J_{\mathbf{k}} = \sum_{\pm\delta_j} (J e^{i\mathbf{k} \cdot \delta_j} - J_z)$  and  $\mathbf{Q} = \mathbf{Q}_0 = (4\pi/3, 0)$ . In both cases, lowering of a magnetic field below  $h_{\text{sat}}$  makes  $(\omega_{\mathbf{k}} - \mu)$  negative at  $\mathbf{k} \approx \pm\mathbf{Q}$ , where  $\mathbf{Q}$  is either  $\mathbf{Q}_i$  or  $\mathbf{Q}_0$ , and drives the

Bose-Einstein condensation (BEC) of magnons. To account for BEC, we introduce two condensates,  $\langle a_{\mathbf{Q}} \rangle = \sqrt{N}\psi_1$  and  $\langle a_{-\mathbf{Q}} \rangle = \sqrt{N}\psi_2$ , where  $\psi_{1,2}$  are complex order parameters. In real space,

$$\langle a_{\mathbf{r}} \rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} \langle a_{\pm\mathbf{k}} \rangle = \psi_1 e^{i\mathbf{Q}\cdot\mathbf{r}} + \psi_2 e^{-i\mathbf{Q}\cdot\mathbf{r}}. \quad (7)$$

The ground state energy, per site, of the *uniform* condensed ground state is expanded in powers of  $\psi_{1,2}$  as

$$E_0/N = -\mu(|\psi_1|^2 + |\psi_2|^2) + \frac{1}{2}\Gamma_1(|\psi_1|^4 + |\psi_2|^4) + \Gamma_2|\psi_1|^2|\psi_2|^2 + \Gamma_3((\bar{\psi}_1\psi_2)^3 + \text{h.c.})\dots \quad (8)$$

where  $\bar{\psi}_j$  denotes complex conjugated of  $\psi_j$ , dots stand for higher order terms, and we omitted a constant term. We verified [18] that higher orders in  $\psi_j$  do not modify our analysis.

Whether the state at  $\mu = 0+$  is co-planar or chiral is decided by the sign of  $\Gamma_1 - \Gamma_2$  [11]. For  $\Gamma_1 < \Gamma_2$ , it is energetically favorable to break  $Z_2$  symmetry between condensates and choose  $\psi_1 \neq 0, \psi_2 = 0$  or vice versa. Parameterizing the condensate as  $\psi_1 = \sqrt{\rho}e^{i\varphi}$ , where  $\rho = \mu/\Gamma_1$ , and using Eq.(A-20), we obtain the cone configuration, Eq.(1). The order parameter manifold of this state is  $O(2) \times Z_2$ , where  $O(2)$  is associated with the phase  $\varphi$ .

When  $\Gamma_1 > \Gamma_2$ , it is energetically favorable to preserve  $Z_2$  symmetry and develop both condensates with equal magnitude  $\rho = \mu/(\Gamma_1 + \Gamma_2)$ , i.e., set  $\psi_1 = \sqrt{\rho}e^{i\theta_1}, \psi_2 = \sqrt{\rho}e^{i\theta_2}$ . This corresponds to co-planar state with the common phase  $\varphi = (\theta_1 + \theta_2)/2$  and the relative phase  $\theta = (\theta_1 - \theta_2)/2$ . The order parameter in this state is given by Eq. (2) with  $\mathbf{Q}$  equal to either  $\mathbf{Q}_i$  ( $J - J'$  model) or  $\mathbf{Q}_0$  (XXZ model). For  $\mathbf{Q} = \mathbf{Q}_i$ , the state is incommensurate co-planar configuration in Fig. 1. The order parameter manifold of this state is  $O(2) \times O(2)$ , where one  $O(2)$  is associated with  $\varphi$  and the other with  $\theta$ . For  $\mathbf{Q} = \mathbf{Q}_0$ , the co-planar order is commensurate. In this case, the symmetry is further reduced by  $\Gamma_3$  term, which is allowed because  $e^{i3\mathbf{Q}_0\cdot\mathbf{r}} = 1$  for all sites  $\mathbf{r}$  of the lattice. This term locks the relative phase of the condensates  $\theta$  to three values, reducing the broken symmetry to  $O(2) \times Z_3$ . For  $\Gamma_3 < 0$ ,  $\theta = \pi\ell/3$ , where  $\ell = 0, 1, 2$ . For  $\Gamma_3 > 0$ ,  $\theta = (2\ell + 1)\pi/6$ . These are  $V$  and  $\Psi$  states in Figs. 1 and 2.

Accidental degeneracy of the isotropic model (3) in the classical limit shows up via  $\Gamma_1^{(0)} = \Gamma_2^{(0)} = 9J$  and  $\Gamma_3^{(0)} = 0$ , where the superscript '0' indicates that these expressions are of zeroth order in  $1/S$ . We now analyze the situation in the presence of anisotropy and quantum fluctuations. We first consider  $J - J'$  model with  $J \neq J'$ , and then XXZ model with  $J_z \neq J$ .

**Phases of the  $J - J'$  model.** We computed  $\Gamma_{1,2}^{(0)}$  for classical spins, but in the presence of the the spatial anisotropy and found that it tilts the balance in favor of the cone phase:  $\Delta\Gamma^{(0)} = \Gamma_2^{(0)} - \Gamma_1^{(0)} = J(1 - J'/J)^2(2 + J'/J)^2 > 0$ . Quantum  $1/S$  corrections, on the other hand, favor the co-

planar state:  $\Delta\Gamma^{(1)} < 0$ . We obtained [18]

$$\Delta\Gamma^{(1)} = \frac{1}{16S} \sum_{\mathbf{k} \in \text{BZ}} \left( \frac{(J_0 + 5J_{\mathbf{k}})^2}{J_0 - J_{\mathbf{k}}} - \frac{(J_0 - 4J_{\mathbf{Q}+\mathbf{k}})^2}{J_{\mathbf{Q}+\mathbf{k}} - J_{\mathbf{Q}}} \right) + \frac{3J}{8S} \approx -\frac{1.6J}{S}. \quad (9)$$

Combining classical and quantum contributions, we find that

$$\Delta\Gamma = \Delta\Gamma^{(0)} + \Delta\Gamma^{(1)} = \frac{9(\delta J)^2}{J} - \frac{1.6J}{S} \quad (10)$$

where, we remind,  $\delta J \equiv J - J'$ . We see that  $\Delta\Gamma < 0$  for  $\delta J < \delta J_c = 0.42J/\sqrt{S}$ , and  $\Delta\Gamma > 0$  for larger  $\delta J$ . The condition  $\Delta\Gamma = 0$  selects the point  $B$  in Fig. 1 [16].

*Split transitions near  $\delta J_c$ .* At  $\mu = 0+$ , the transition between incommensurate planar and cone phases is first order with no hysteresis. We now analyze how this transition occurs at a finite positive  $\mu \neq 0$ . We depart from the cone state to the right of point  $B$  in Fig. 1 and move to smaller  $\delta J$ . Suppose that the condensate in the cone state has momentum  $+\mathbf{Q}_i$ . Then Goldstone spin-wave mode is at  $\mathbf{k} = \mathbf{Q}_i$ , while excitations near  $\mathbf{k} = -\mathbf{Q}_i$  have a finite gap. We computed the excitation spectrum  $\omega_{\mathbf{k}}^{(1)}$  with quantum  $1/S$  corrections and found [18] that near  $\mathbf{k} \approx -\mathbf{Q}_i$

$$\omega_{\mathbf{k}}^{(1)} \approx \frac{3J}{4} [(k_x + \bar{Q}_i)^2 + k_y^2 + \epsilon_{\min}], \quad (11)$$

$$\epsilon_{\min} = \frac{12\mu}{h_{\text{sat}}J^2} \left[ (\delta J)^2 - (\delta J_c)^2 \left( 1 + \frac{\mu}{h_{\text{sat}}} \right) \right], \quad (12)$$

where  $\bar{Q}_i = Q_i + (4\pi/3 - Q_i)(3\mu/h_{\text{sat}}) \approx Q_i + 1.45\mu/(h_{\text{sat}}\sqrt{S})$ . The cone state becomes unstable at  $\epsilon_{\min} = 0$ , i.e., at  $\delta J_{c3} \approx \delta J_c(1 + \mu/(2h_{\text{sat}}))$ , and gives rise to magnon condensation with momentum  $(-\bar{Q}_i, 0)$ , which is *different* from  $-\mathbf{Q}_i$ . The condensation of magnons with  $(-\bar{Q}_i, 0)$  then gives rise to a secondary cone order, with momentum not related by symmetry to that of the primary cone order. The resulting spin configuration is a double cone with  $O(2) \times O(2) \times Z_2$  order parameter manifold. The primary condensate sets the transverse component of  $\langle \mathbf{S}_{\mathbf{r}}^{\perp} \rangle = \langle S_{\mathbf{r}}^x + iS_{\mathbf{r}}^y \rangle$  to be  $\exp[i\mathbf{Q}_i \cdot \mathbf{r} + i\theta_1]$  and the second condensate adds  $\exp[-i\bar{\mathbf{Q}}_i \cdot \mathbf{r} + i\theta_2]$ .

At smaller  $\delta J \leq \delta J_{c3}$  the position of the minimum in  $\omega_{\mathbf{k}}^{(1)}$  in (11) evolves and drifts towards  $-\mathbf{Q}_i$ . Once it reaches  $-\mathbf{Q}_i$ , at  $\delta J = \delta J_{c2}$ , the two cone configurations interfere constructively and give rise to an incommensurate co-planar state. Critical  $\delta J_{c2}$  can be estimated by requiring that  $\omega_{\mathbf{k}}^{(1)} = 0$  at  $\mathbf{k} = -\mathbf{Q}_i$ . This yields  $\delta J_{c2} = \delta J_{c3}(1 - O(\mu/h_{\text{sat}})) < \delta J_{c3}$ . We see therefore that the transformation from a cone to an incommensurate co-planar state at a finite  $\mu$  (i.e., at  $h \leq h_{\text{sat}}$ ) occurs via two transitions at  $\delta J_{c2}$  and  $\delta J_{c3}$  and involves an intermediate double cone phase (Fig. 1).

*Instability of the  $V$  phase.* We now return to Eq. (8) and consider the transition between the  $V$  phase and the incommensurate co-planar phase. At  $\mu = 0+$ , this transition holds at infinitesimally small  $\delta J$  (point  $A$  in Fig. 1). We

show that at a finite  $\mu$ , the  $V$  phase survives up to a finite  $\delta J_{c1} \sim (J/\sqrt{S})(\mu/h_{\text{sat}})$ . The argument is that in the  $V$  phase  $\mathbf{Q} = \mathbf{Q}_0$  is commensurate and  $\Gamma_3$  term in Eq. (8) is allowed. We recall that at  $\delta J = 0$  and for classical spins  $\Gamma_3 = 0$ . We computed the classical contribution to  $\Gamma_3$  at  $\delta J > 0$  and the contribution due to quantum fluctuations at  $\delta J = 0$ . We found [18] that the classical contribution vanishes, but the quantum contribution is finite to order  $1/S^2$  and makes  $\Gamma_3$  negative:

$$\Gamma_3 = \frac{3}{32S^2} \sum_{\mathbf{k} \in \text{BZ}} \left( \frac{(5J_{\mathbf{k}} + J_0)(5J_{\mathbf{Q}+\mathbf{k}} + J_0)J_{\mathbf{Q}-\mathbf{k}}}{(J_0 - J_{\mathbf{k}})(J_0 - J_{\mathbf{Q}+\mathbf{k}})} - \frac{(5J_{\mathbf{k}} + J_0)(J_{\mathbf{k}} + J_0)}{2(J_0 - J_{\mathbf{k}})} \right) + \frac{3J_0}{64S^2} \approx -\frac{0.69J}{S^2} \quad (13)$$

Because  $\Gamma_3 < 0$ , the  $V$  phase has extra negative energy compared to incommensurate phases, and one needs a finite  $\delta J$  to overcome this energy difference.

We now argue that the transition at  $\delta J_{c1}$  belongs to the special class of CIT. To see this, we allow for spatially non-uniform configurations of the condensate  $\psi_{1,2}(\mathbf{r})$ . This adds spatial gradient terms to (4): the isotropic term  $\mathcal{H}_0$  produces conventional quadratic in gradient contribution  $\propto \rho(\partial_x \theta)^2$ , while  $\delta \mathcal{H}_{\text{anis}}$  adds a *linear* gradient term  $\propto \rho S \delta J \partial_x (\theta_1 - \theta_2)$ . Combining these two classical contributions with the quantum  $\Gamma_3$  term in (8), we obtain the energy density for the relative phase  $\theta = (\theta_1 - \theta_2)/2$ :

$$\mathcal{E}_\theta = \frac{3JS^2\mu}{4h_{\text{sat}}}(\partial_x \theta)^2 + \frac{\sqrt{3}\delta JS^2\mu}{h_{\text{sat}}}\partial_x \theta + S \frac{(\Gamma_3 S^2)}{4} \frac{\mu^3}{h_{\text{sat}}^3} \cos[6\theta] \quad (14)$$

Eq. (14) is of standard sine-Gordon form, which allows us to borrow the results from [14]: the equilibrium value of  $\theta$  shifts from the commensurate  $\theta = \pi\ell/3$  in the  $V$  phase to an incommensurate value when the coefficient of the linear gradient term in (14) exceeds the geometric mean of the coefficients of two other terms in (14). Using Eq. (14) we find that CIT occurs at  $\delta J_{c1} = 1.17(J/\sqrt{S})(\mu/h_{\text{sat}}) = 0.13\mu/S^{3/2}$  (line AC in Fig. 1). At  $\delta J > \delta J_{c1}$ ,  $\theta$  acquires linear dependence on  $x$ :  $\theta = \tilde{Q}x + \tilde{\theta}$ . In this situation, the spin configuration becomes incommensurate but remains co-planar (Fig. 1).

The critical  $\delta J_{c1}$  for the CIT has to be compared with  $\delta J_{\text{sw}}$  at which spin-wave excitations in the  $V$  phase soften. We computed spin-wave velocity with quantum  $1/S$  corrections and found that it does go down with increasing  $\delta J$  but vanishes only at  $\delta J_{\text{sw}} \sim (J/\sqrt{S})(\mu/h_{\text{sat}})^{1/2} \gg \delta J_{c1}$ . This implies that the spin-wave velocity remains finite across the CIT.

**Phases of  $\mathcal{H}_{\text{XXZ}}$ .** For the XXZ model with exchange anisotropy,  $J$  and  $J'$  remain equal, but  $J_z < J_\perp = J$  on all bonds. We verified [18] that  $\mathbf{Q}$  remains commensurate for all  $J_z/J \leq 1$ , i.e.,  $\mathbf{Q} = \mathbf{Q}_0 = (4\pi/3, 0)$ . In this situation, we found  $\Gamma_2^{(0)} - \Gamma_1^{(0)} = -J_{\mathbf{Q}}(1 - J_z/J) = 3J\Delta$ . Quantum corrections to  $\Gamma_1$  and  $\Gamma_2$  are determined within the same isotropic model (3) and are given by (10). Using this, we immediately find that the ground state of the quantum XXZ model is coplanar for  $\Delta \leq \Delta_{c2} = 0.53/S$  and is a cone for  $\Delta > \Delta_{c2}$ .

The transition between co-planar and cone states near  $\Delta_{c2}$  remains first-order for a finite  $\mu > 0$ , i.e., no intermediate double spiral state appears. This is the consequence of the fact that  $\mathbf{Q} = \mathbf{Q}_0$  remains commensurate. Still, the transformation from the  $V$  phase to the cone phase does involve a new intermediate state, which comes about due to the change of sign of  $\Gamma_3$ . Exchange anisotropy  $\Delta$  gives rise to a positive  $\Gamma_3$  to order  $1/S$ :  $\Gamma_3^{(1)} = J(1 + 2J_z/J)(1 - J_z/J)/(2S) \approx 3J\Delta/(2S)$  (see [18] for details). At the same time the quantum corrections give rise to *negative*  $\Gamma_3$  to order  $1/S^2$  already at  $\Delta = 0$ , see (A-43). Combining the two, we find that

$$\Gamma_3 = \Gamma_3^{(1)} + \Gamma_3^{(2)} = \frac{3J\Delta}{2S} - \frac{0.69J}{S^2}. \quad (15)$$

changes sign at  $\Delta_{c1} = 0.45/S < \Delta_{c2} = 0.53/S$ . At smaller  $\Delta < \Delta_{c1}$ ,  $\Gamma_3 < 0$ , and the spin configuration is the  $V$  state (the energy is minimized by setting  $\cos 6\theta = 1$ , see (8)). However, in the interval  $\Delta_{c1} < \Delta < \Delta_{c2}$ ,  $\Gamma_3 > 0$  becomes positive. The energy is now minimized by  $\cos 6\theta = -1$ , which corresponds to the  $\Psi$  state in Fig. 2. The transition is highly unconventional symmetry-wise because the order parameter manifold is  $O(2) \times Z_3$  in both phases, but extends to a larger  $O(2) \times O(2)$  symmetry at the transition point.

We present the phase diagram of XXZ model in Fig. 2. A very similar phase diagram has been recently obtained in the numerical cluster mean-field analysis of the  $S = 1/2$  XXZ model [17].

To summarize, in this paper we considered anisotropic 2D Heisenberg antiferromagnets on a triangular lattice in a high magnetic field close to the saturation. We analyzed the cases of spatially anisotropic interactions, like in  $\text{Cs}_2\text{CuCl}_4$  and  $\text{Cs}_2\text{CuBr}_4$  and of exchange anisotropy, as in  $\text{Ba}_3\text{CoSb}_2\text{O}_9$ . We showed that the phase diagram in field/anisotropy plane is quite rich due to competition between classical and quantum orders, which favor non-coplanar and co-planar states, respectively. This competition leads to multiple transitions and highly non-trivial intermediate phases, including a novel double cone state. We demonstrated that one of the transition in each of the two cases studied is of CIT type and is not accompanied by softening of spin-wave excitations.

The analysis of this paper can be easily extended to quasi-2D layered systems, with inter-layer antiferromagnetic interaction  $0 < J'' \ll J$ . This additional exchange interaction leads to the staggering of coplanar spin configurations, of either  $V$  or  $\Psi$  kind, between the adjacent layers, as can easily be seen by treating  $\varphi \rightarrow \varphi_z$  in Eq.(2) as layer-dependent variable with discrete index  $z$ . One then immediately finds that  $J'' \sum_{\mathbf{r}, z} \tilde{S}_{\mathbf{r}, z} \cdot \tilde{S}_{\mathbf{r}, z+1}$  is minimized by  $\varphi_z = \varphi + \pi z$ , in agreement with earlier spin-wave [19] and Monte Carlo [9] studies.

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## Supplementary Information for “Phases of triangular lattice antiferromagnet near saturation”

Oleg A. Starykh, Wen Jin, and Andrey V. Chubukov

Here we present technical details of calculations reported in the manuscript. All calculations were carried out in one-sublattice and in three-sublattice basis, and led to identical results. For definiteness, we present the details of calculations in the one-sublattice basis.

### THE HAMILTONIAN AND THE EXPANSION IN BOSONS

We consider Heisenberg Hamiltonian of 2D triangular lattice (Eq. (3) of the main text), and expand it to sixth order in Holstein Primakoff bosons around the ferromagnetic state, which holds at  $h > h_{sat}$ . We then move to fields below the saturation value by introducing magnon condensates and using the technique of dilute Bose-gas expansion.

The Hamiltonian in terms of Holstein Primakoff bosons has the form

$$\mathcal{H} = \mathcal{H}^{(2)} + \mathcal{H}^{(4)} + \mathcal{H}^{(6)},$$

$$\mathcal{H}^{(2)} = \sum_{\mathbf{k}} (\omega_{\mathbf{k}} - \mu) a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}}, \quad (\text{A-1})$$

$$\mathcal{H}^{(4)} = \frac{1}{2N} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} V_{\mathbf{q}}(\mathbf{k}, \mathbf{k}') a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}'-\mathbf{q}}^{\dagger} a_{\mathbf{k}'} a_{\mathbf{k}}, \quad (\text{A-2})$$

$$\mathcal{H}^{(6)} = \frac{1}{16SN^2} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{k}'', \mathbf{q}, \mathbf{p}} U_{\mathbf{q}, \mathbf{p}}(\mathbf{k}, \mathbf{k}', \mathbf{k}'') a_{\mathbf{k}+\mathbf{q}+\mathbf{p}}^{\dagger} a_{\mathbf{k}'-\mathbf{q}}^{\dagger} a_{\mathbf{k}''-\mathbf{p}}^{\dagger} a_{\mathbf{k}''} a_{\mathbf{k}'} a_{\mathbf{k}}. \quad (\text{A-3})$$

Here,  $a, a^{\dagger}$  are boson operators,  $\omega_{\mathbf{k}}$  is the magnon dispersion,  $\mu = h_{sat} - h$  is the chemical potential, and  $V_{\mathbf{q}}(\mathbf{k}, \mathbf{k}')$ ,  $U_{\mathbf{q}, \mathbf{p}}(\mathbf{k}, \mathbf{k}', \mathbf{k}'')$  are 2- and 3-body interaction potentials which we list below separately for isotropic and anisotropic models. Both  $\omega_{\mathbf{k}}$  and  $h_{sat}$  are of order  $S$ , and we consider  $\mu$  also of order  $S$ .

#### Isotropic Heisenberg Model

In the isotropic case

$$\omega_{\mathbf{k}} = S(J_{\mathbf{k}} - J_{\mathbf{Q}}), \quad (\text{A-4})$$

$$V_{\mathbf{q}}(\mathbf{k}, \mathbf{k}') = \frac{1}{2} [J_{\mathbf{k}-\mathbf{k}'+\mathbf{q}} + J_{\mathbf{q}} - \frac{1}{2}(J_{\mathbf{k}+\mathbf{q}} + J_{\mathbf{k}'-\mathbf{q}} + J_{\mathbf{k}} + J_{\mathbf{k}'})], \quad (\text{A-5})$$

$$U_{\mathbf{q}, \mathbf{p}}(\mathbf{k}, \mathbf{k}', \mathbf{k}'') = \frac{1}{9} \left( J_{\mathbf{k}+\mathbf{q}} + J_{\mathbf{k}''+\mathbf{q}} + J_{\mathbf{k}+\mathbf{k}''-\mathbf{k}'+\mathbf{q}} + J_{\mathbf{k}+\mathbf{p}} + J_{\mathbf{k}'+\mathbf{p}} + J_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''+\mathbf{p}} \right. \\ \left. + J_{\mathbf{k}'+\mathbf{k}''-\mathbf{k}-\mathbf{q}-\mathbf{p}} + J_{\mathbf{k}''-\mathbf{q}-\mathbf{p}} + J_{\mathbf{k}'-\mathbf{q}-\mathbf{p}} \right) \\ - \frac{1}{6} \left( J_{\mathbf{k}+\mathbf{q}+\mathbf{p}} + J_{\mathbf{k}'-\mathbf{q}} + J_{\mathbf{k}''-\mathbf{p}} + J_{\mathbf{k}} + J_{\mathbf{k}'} + J_{\mathbf{k}''} \right). \quad (\text{A-6})$$

where  $J_{\mathbf{k}} = 2J(\cos[k_x] + 2\cos[\frac{k_x}{2}]\cos[\frac{\sqrt{3}k_y}{2}])$ , with its minimum  $J_{\mathbf{Q}}$  at  $\mathbf{Q} = (Q_0, 0)$ , and  $Q_0 = 4\pi/3$ .

#### Anisotropic $J$ - $J'$ Model

In this model,  $\omega_{\mathbf{k}}$ ,  $V_{\mathbf{q}}(\mathbf{k}, \mathbf{k}')$ , and  $U_{\mathbf{q}, \mathbf{p}}(\mathbf{k}, \mathbf{k}', \mathbf{k}'')$  are all in the same form as  $J_{\mathbf{k}}$  above, except replacing all  $J_{\mathbf{k}}$  with  $\tilde{J}_{\mathbf{k}}$ , where  $\tilde{J}_{\mathbf{k}} = 2(J\cos[k_x] + 2J'\cos[\frac{k_x}{2}]\cos[\frac{\sqrt{3}k_y}{2}])$ .  $\tilde{J}_{\mathbf{k}}$  has minimum  $\tilde{J}_{\mathbf{Q}}$  at  $\mathbf{Q} = (Q_i, 0)$ , and  $Q_i = 2\cos^{-1}[-J'/2J]$ .

#### XXZ Model

In this model,  $\omega_{\mathbf{k}}$  is same as Eq.(A-4), and  $U_{\mathbf{q}, \mathbf{p}}(\mathbf{k}, \mathbf{k}', \mathbf{k}'')$  is same as Eq.(A-6). The difference comes from  $V_{\mathbf{q}}(\mathbf{k}, \mathbf{k}')$ , which now contains the exchange anisotropy in the  $z$  direction:

$$V_{\mathbf{q}}(\mathbf{k}, \mathbf{k}') = \frac{1}{2} \left[ J_{\mathbf{k}-\mathbf{k}'+\mathbf{q}}^z + J_{\mathbf{q}}^z - \frac{1}{2}(J_{\mathbf{k}+\mathbf{q}} + J_{\mathbf{k}'-\mathbf{q}} + J_{\mathbf{k}} + J_{\mathbf{k}'} \right], \quad (\text{A-7})$$

where  $J_{\mathbf{k}}^z = 2J^z(\cos[k_x] + 2\cos[\frac{k_x}{2}]\cos[\frac{\sqrt{3}k_y}{2}])$ . The minimum of  $J_{\mathbf{k}}^z$  is at  $\mathbf{k} = (Q_0, 0)$ .

### CALCULATION OF $\Gamma_1, \Gamma_2, \Gamma_3$

We follow [1] and split magnon operators into condensate and non-condensate fractions as

$$a_{\mathbf{k}} = \sqrt{N}\psi_1\delta_{\mathbf{k},\mathbf{Q}} + \sqrt{N}\psi_2\delta_{\mathbf{k},-\mathbf{Q}} + \tilde{a}_{\mathbf{k}}, \quad (\text{A-8})$$

where  $\psi_{1,2}$  describe condensates at momenta  $\mathbf{k} = \mathbf{Q}$  and  $\mathbf{k} = -\mathbf{Q}$ , and  $\tilde{a}_{\mathbf{k}}$  describes non-condensate magnons. The ground state energy density reads

$$E_0/N = -\mu(|\psi_1|^2 + |\psi_2|^2) + \frac{1}{2}\Gamma_1(|\psi_1|^4 + |\psi_2|^4) + \Gamma_2|\psi_1|^2|\psi_2|^2 + \Gamma_3((\bar{\psi}_1\psi_2)^3 + \text{h.c.}) \quad (\text{A-9})$$

The classical expressions for  $\Gamma_1$  and  $\Gamma_2$  (the ones at order  $1/S^0$ ) are obtained by neglecting all non-condensate modes and are shown schematically in Fig.A-1. These contributions are related to potential  $V_{\mathbf{q}}(\mathbf{k}, \mathbf{k}')$  via

$$\Gamma_1^{(0)} = V_0(\mathbf{Q}, \mathbf{Q}), \quad (\text{A-10})$$

$$\Gamma_2^{(0)} = V_0(\mathbf{Q}, -\mathbf{Q}) + V_{2\mathbf{Q}}(-\mathbf{Q}, \mathbf{Q}). \quad (\text{A-11})$$

The classical expression for  $\Gamma_3$  (at order  $1/S$ ) is shown schematically in Fig.A-1 and it is related to potential  $V_{\mathbf{q}}(\mathbf{k}, \mathbf{k}')$  and  $U_{\mathbf{q},\mathbf{p}}(\mathbf{k}, \mathbf{k}', \mathbf{k}'')$  via

$$\Gamma_3^{(1)} = \frac{U_{2\mathbf{Q},2\mathbf{Q}}(\mathbf{Q}, \mathbf{Q}, \mathbf{Q})}{16S} - \frac{[V_{2\mathbf{Q}}(\mathbf{Q}, \mathbf{Q})]^2}{\omega_{3\mathbf{Q}}}. \quad (\text{A-12})$$

Here the first term comes directly from the Hamiltonian (A-3), and the second one originates from the condensate  $\psi_0 \equiv \langle \tilde{a}_0 \rangle \neq 0$ , which is induced at the momentum  $\mathbf{k} = 3\mathbf{Q} = 0$  in the case of *commensurate* ordering at wave vector  $\mathbf{Q} = (4\pi/3_0, 0)$ . This novel condensate adds the term  $|\psi_0|^2\omega_0 + V_{2\mathbf{Q}}(\mathbf{Q}, \mathbf{Q})[\psi_0(\bar{\psi}_1\psi_2^2 + \psi_1^2\bar{\psi}_2) + \text{h.c.}]$  to the ground state energy. Minimizing this extra energy contribution, we find the expression for  $\psi_0$

$$\psi_0 = -\frac{V_{2\mathbf{Q}}(\mathbf{Q}, \mathbf{Q})}{\omega_0}(\bar{\psi}_1\psi_2^2 + \psi_1^2\bar{\psi}_2) = \frac{1}{4S}(\bar{\psi}_1\psi_2^2 + \psi_1^2\bar{\psi}_2). \quad (\text{A-13})$$

It is important to keep in mind that this result is derived for  $\mathbf{Q} = (4\pi/3_0, 0)$ , when  $e^{i3\mathbf{Q}\cdot\mathbf{r}} = 1$  for all sites of the triangular lattice  $\mathbf{r}$ .

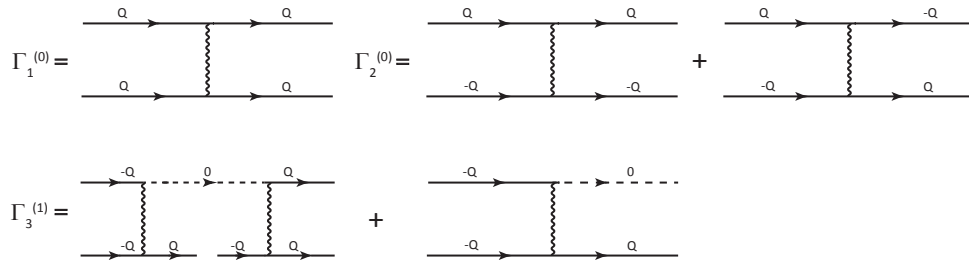


FIG. A-1. Diagrams for  $\Gamma_1, \Gamma_2$  and  $\Gamma_3$  in the classical limit.

The expressions for  $\Gamma_1^{(0)}, \Gamma_2^{(0)}$ , and  $\Gamma_3^{(1)}$  are different in the isotropic case and in the two anisotropic cases.

For the isotropic model,

$$\begin{aligned} \Gamma_1^{(0)} &= J_0 - J_{\mathbf{Q}}, \Gamma_2^{(0)} = J_0 + J_{2\mathbf{Q}} - J_{\mathbf{Q}}, \\ \Gamma_3^{(1)} &= 0. \end{aligned} \quad (\text{A-14})$$

For  $J - J'$  model,

$$\begin{aligned}\Gamma_2^{(0)} - \Gamma_1^{(0)} &= \tilde{J}_{2\mathbf{Q}} - \tilde{J}_{\mathbf{Q}} = J(2 + \frac{J'}{J})^2(1 - \frac{J'}{J})^2 \approx \frac{9(\delta J)^2}{J}, \\ \Gamma_3^{(1)} &= 0.\end{aligned}\tag{A-15}$$

For XXZ model,

$$\begin{aligned}\Gamma_2^{(0)} - \Gamma_1^{(0)} &= J_{2\mathbf{Q}}^z - J_{\mathbf{Q}} = 3J\Delta, \\ \Gamma_3^{(1)} &= \frac{J_0 - J_{\mathbf{Q}}}{16S} - \frac{(4J_{\mathbf{Q}}^z - 3J_{\mathbf{Q}} - J_0)^2}{16S(J_0 - J_{\mathbf{Q}})} = \frac{J}{2S}(1 + \frac{J_z}{J})(1 - \frac{J_z}{J}) \approx \frac{3J\Delta}{2S}.\end{aligned}\tag{A-16}$$

### QUANTUM CORRECTIONS TO $\Gamma_1, \Gamma_2, \Gamma_3$

In this section, we compute quantum corrections to  $\Gamma_1, \Gamma_2, \Gamma_3$ . Because these corrections already contain extra factor of  $1/S$ , they can be calculated by neglecting anisotropy. Quantum corrections to  $\Gamma_{1,2}$  are of order  $1/S$ , and quantum corrections to  $\Gamma_3$  are of order  $(1/S)^2$ . In both cases, quantum term has extra factor  $1/S$  compared to classical results. Each quantum correction is a sum of the two terms – one comes from normal ordering of Holstein-Primakoff bosons, and the other from second and third-order terms in the perturbation expansion in  $1/S$ .

#### Corrections from normal ordering

The Holstein-Primakoff transformation

$$S^z(r) = S - a_{\mathbf{r}}^+ a_{\mathbf{r}}, S^+ = \sqrt{2S - a_{\mathbf{r}}^+ a_{\mathbf{r}}} a_{\mathbf{r}}, S^- = \sqrt{2S} a_{\mathbf{r}}^+ \sqrt{2S - a_{\mathbf{r}}^+ a_{\mathbf{r}}}\tag{A-17}$$

contains the square-root  $\sqrt{2S - a_{\mathbf{r}}^+ a_{\mathbf{r}}}$ , which needs to be expanded in the *normal-ordered* form to perform dilute gas analysis (all  $a_{\mathbf{r}}^+$  have to stand to the left of  $a_{\mathbf{r}}$ ). Because  $a_{\mathbf{r}}^+ a_{\mathbf{r}} = a_{\mathbf{r}} a_{\mathbf{r}}^+ - 1$ , i.e.,  $(a_{\mathbf{r}}^+ a_{\mathbf{r}})^2 = a_{\mathbf{r}}^+ a_{\mathbf{r}}^+ a_{\mathbf{r}} a_{\mathbf{r}} + a_{\mathbf{r}}^+ a_{\mathbf{r}}$ , etc, the prefactors in this *normal-ordering* are not simply powers of  $1/S$  but rather contain series of  $1/S$  terms. To order  $1/S^3$  we have

$$S_{\mathbf{r}}^- = \sqrt{2S} a_{\mathbf{r}}^+ \left\{ 1 - \frac{1}{4S} \left( 1 + \frac{1}{8S} + \frac{1}{32S^2} \right) a_{\mathbf{r}}^+ a_{\mathbf{r}} - \frac{1}{32S^2} \left( 1 + \frac{3}{4S} \right) a_{\mathbf{r}}^+ a_{\mathbf{r}}^+ a_{\mathbf{r}} a_{\mathbf{r}} - \frac{a_{\mathbf{r}}^+ a_{\mathbf{r}}^+ a_{\mathbf{r}}^+ a_{\mathbf{r}} a_{\mathbf{r}} a_{\mathbf{r}}}{128S^3} + O(1/S^4) \right\}$$

The  $1/S$  corrections to the prefactors modify Eqs.(A-2) and (A-3) to

$$\delta\mathcal{H}^{(4)} = -\frac{J}{32S} \sum_{\mathbf{r}, \delta} (a_{\mathbf{r}}^{\dagger} a_{\mathbf{r}+\delta}^{\dagger} a_{\mathbf{r}} a_{\mathbf{r}+\delta} + \text{h.c.}),\tag{A-18}$$

$$\delta\mathcal{H}^{(6)} = \frac{J}{128S^2} \sum_{\mathbf{r}, \delta} (a_{\mathbf{r}}^{\dagger} a_{\mathbf{r}+\delta}^{\dagger} a_{\mathbf{r}+\delta}^{\dagger} a_{\mathbf{r}} a_{\mathbf{r}} a_{\mathbf{r}+\delta} + \text{h.c.}) - \frac{3J}{128S^2} \sum_{\mathbf{r}, \delta} (a_{\mathbf{r}}^{\dagger} a_{\mathbf{r}}^{\dagger} a_{\mathbf{r}+\delta}^{\dagger} a_{\mathbf{r}} a_{\mathbf{r}} a_{\mathbf{r}} + \text{h.c.}).\tag{A-19}$$

Substituting the form of the condensate in real space

$$\langle a_{\mathbf{r}} \rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}} \langle a_{\pm\mathbf{Q}} \rangle = \psi_1 e^{i\mathbf{Q} \cdot \mathbf{r}} + \psi_2 e^{-i\mathbf{Q} \cdot \mathbf{r}}.\tag{A-20}$$

we obtain  $1/S$  corrections to classical expressions for  $\Gamma_{1,2,3}$ :

$$\begin{aligned}\Delta\Gamma_a^{(1)} &= \Gamma_{2a}^{(1)} - \Gamma_{1a}^{(1)} = \left(-\frac{J_{\mathbf{Q}}}{4S}\right) - \left(-\frac{J_{\mathbf{Q}}}{8S}\right) = \frac{3J}{8S}, \\ \Gamma_{3a}^{(2)} &= \frac{5J_0}{128S^2} + \frac{J_0}{128S^2} = \frac{9J}{32S^2}.\end{aligned}\tag{A-21}$$



### Corrections from quantum fluctuations

To find quantum corrections to parameters  $\Gamma_{1,2,3}$ , we evaluate corrections to the ground state energy density  $\delta E$  from non condensed modes  $\tilde{a}_{\mathbf{k}}$  in (A-8) in perturbation theory up to third order and obtain the correction to the ground state energy density  $\Delta E$  to sixth order in the condensates  $\psi_1$  and  $\psi_2$ . The prefactors for the  $\psi^4$  and  $\psi^6$  term in  $\Delta E$  yield quantum corrections to interaction parameters  $\Gamma_{1,2,3}$ .

Quite generally, under perturbation  $H_i$ , the partition function is

$$Z = \int \prod_{\mathbf{k}} da_{\mathbf{k}}^\dagger da_{\mathbf{k}} e^{\int_0^\beta d\tau (L_0 - H_i)} = Z_0 \frac{\int \prod_{\mathbf{k}} da_{\mathbf{k}}^\dagger da_{\mathbf{k}} e^{\int_0^\beta d\tau (L_0 - H_i)}}{\int \prod_{\mathbf{k}} da_{\mathbf{k}}^\dagger da_{\mathbf{k}} e^{\int_0^\beta L_0}} \equiv Z_0 \langle e^{-\int_0^\beta H_i} \rangle_0. \quad (\text{A-22})$$

Here  $L_0 = \sum_{\mathbf{k}} (a_{\mathbf{k}}^\dagger \frac{\partial}{\partial \tau} a_{\mathbf{k}}) - \mathcal{H}^{(2)}$  represents Lagrangian of non-interacting magnons described by the quadratic Hamiltonian (6), and  $\beta = 1/T$ . The internal energy density is

$$E = -\frac{\partial \ln Z}{\partial \beta} \approx -\frac{\partial \ln Z_0}{\partial \beta} - \frac{\partial (\beta \ln \langle e^{-H_i} \rangle)}{\partial \beta} = E_0 + \Delta E \quad (\text{A-23})$$

The correction term  $\Delta E$  is represented by the standard cumulant expansion, which involves only connected averages of the perturbation  $H_i$

$$\Delta E = \langle H_i \rangle_0 - \frac{1}{2!} \langle \int_\tau H_i^2 \rangle_0 + \frac{1}{3!} \langle \int_\tau \int_{\tau'} H_i^3 \rangle_0 + \dots \quad (\text{A-24})$$

In the zero-temperature limit, in which all our calculations are done,  $E = E_0 + \Delta E$  determines the ground state energy. Integration over relative times  $\tau, \tau' \dots$  ensures conservation of frequencies in the internal vertices of the diagrams. The role of the perturbation  $H_i$  is played by interacting Hamiltonians (A-2), (A-3) expressed in terms of condensates  $\psi_{1,2}$  and non-condensed magnons  $\tilde{a}_{\mathbf{k}}$  after the substitution (A-8). We remind that the averaging is over the free-boson Hamiltonian for isotropic system at  $h = h_{sat}$ .

### Quantum corrections to $\Gamma_{1,2}$

Quantum corrections to  $\Gamma_{1,2}$  are of order  $1/S$ , and to get them we only need the fourth-order term in bosons (A-2):

$$\mathcal{H}_{i,\mathbf{k}} = \sum_{\mathbf{k}} \left[ \left( \frac{1}{2} V_{\mathbf{k}}(\mathbf{Q}, \mathbf{Q}) \psi_1^2 a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger + V_{\mathbf{k}}(\mathbf{Q}, -\mathbf{Q}) \psi_1 \psi_2 a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger + \frac{1}{2} V_{\mathbf{k}}(-\mathbf{Q}, -\mathbf{Q}) \psi_2^2 a_{-\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger \right) + \text{h.c.} \right], \quad (\text{A-25})$$

where  $V_{\mathbf{q}}(k, k')$  is defined in Eq.(A-5). The first-order correction to the energy density obviously vanishes, and the second-order perturbative correction yields

$$\begin{aligned} \Delta E &= -\frac{1}{2} \sum_{\mathbf{k}, \mathbf{q}} \langle \mathcal{H}_{i,\mathbf{k}} \cdot \mathcal{H}_{i,\mathbf{q}} \rangle_0 \\ &= -\sum_{\mathbf{k}, \mathbf{q}} \left[ \frac{1}{4} |\psi_1|^4 V_{\mathbf{k}}(\mathbf{Q}, \mathbf{Q}) V_{\mathbf{q}}(\mathbf{Q}, \mathbf{Q}) \langle a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger a_{\mathbf{Q}+\mathbf{q}} a_{\mathbf{Q}-\mathbf{q}} \rangle_0 \right. \\ &\quad + \frac{1}{4} |\psi_2|^4 V_{\mathbf{k}}(-\mathbf{Q}, -\mathbf{Q}) V_{\mathbf{q}}(-\mathbf{Q}, -\mathbf{Q}) \langle a_{-\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger a_{-\mathbf{Q}+\mathbf{q}} a_{-\mathbf{Q}-\mathbf{q}} \rangle_0 \\ &\quad \left. + \frac{1}{2} |\psi_1|^2 |\psi_2|^2 V_{\mathbf{k}}(\mathbf{Q}, -\mathbf{Q}) V_{\mathbf{q}}(\mathbf{Q}, -\mathbf{Q}) \langle a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger a_{\mathbf{Q}+\mathbf{q}} a_{-\mathbf{Q}-\mathbf{q}} \rangle_0 \right]. \quad (\text{A-26}) \end{aligned}$$

By Wick's theorem,

$$\langle a_{k_1}^\dagger a_{k_2}^\dagger a_{k_3} a_{k_4} \rangle_0 = \langle a_{k_1}^\dagger a_{k_3} \rangle_0 \langle a_{k_2}^\dagger a_{k_4} \rangle_0 + \langle a_{k_1}^\dagger a_{k_4} \rangle_0 \langle a_{k_2}^\dagger a_{k_3} \rangle_0. \quad (\text{A-27})$$

where the pair average is [2]

$$\langle a_{k_1}^\dagger a_{k_2} \rangle_0 = -\delta_{k_1, k_2} G_0(k_1), \quad (\text{A-28})$$

and  $G_0(k) \equiv G_0(k, \epsilon)$  is the free boson Green's function

$$G_0(k) = (i\omega - \epsilon_k)^{-1}, \quad (\text{A-29})$$

Utilizing the properties of (A-27) and (A-28), we obtain the terms in the form

$$\sum_{\mathbf{k}, \mathbf{q}} V_{\mathbf{k}}(\mathbf{Q}, \mathbf{Q}) V_{\mathbf{q}}(\mathbf{Q}, \mathbf{Q}) \langle a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger a_{\mathbf{Q}+\mathbf{q}} a_{\mathbf{Q}-\mathbf{q}} \rangle_0 = \sum_{\mathbf{k}, \omega} \frac{2V_{\mathbf{k}}^2(\mathbf{Q}, \mathbf{Q})}{(i\omega - \epsilon_{\mathbf{Q}+\mathbf{k}})(i\omega - \epsilon_{\mathbf{Q}-\mathbf{k}})} \quad (\text{A-30})$$

Using

$$\text{with } \sum_{\omega} \frac{1}{(i\omega - \epsilon_1)(i\omega - \epsilon_2)} = \int \frac{d\omega}{2\pi} \frac{1}{(i\omega - \epsilon_1)(i\omega - \epsilon_2)} = \frac{1}{\epsilon_1 + \epsilon_2} \quad (\text{A-31})$$

and collecting prefactors we obtain the corrections to  $\Gamma_{1,2}$  in the form

$$\begin{aligned} \Gamma_{1b}^{(1)} &= - \sum_{\mathbf{k}} \frac{V_{\mathbf{k}}^2(\mathbf{Q}, \mathbf{Q})}{\omega_{\mathbf{Q}+\mathbf{k}} + \omega_{\mathbf{Q}-\mathbf{k}}} = - \frac{1}{16S} \sum_{\mathbf{k}} \frac{(J_0 + 5J_{\mathbf{k}})^2}{J_0 - J_{\mathbf{k}}}, \\ \Gamma_{2b}^{(1)} &= - \sum_{\mathbf{k}} \frac{V_{\mathbf{k}}^2(\mathbf{Q}, -\mathbf{Q})}{\omega_{\mathbf{Q}+\mathbf{k}}} = - \frac{1}{16S} \sum_{\mathbf{k}} \frac{(J_0 - 4J_{\mathbf{Q}+\mathbf{k}})^2}{J_{\mathbf{Q}+\mathbf{k}} - J_{\mathbf{k}}}. \end{aligned} \quad (\text{A-32})$$

These corrections can be equally obtained diagrammatically, by evaluating second-order corrections to  $\phi^4$  vertices, as in Fig. A-2.

Each of the two integrals above is logarithmically divergent, but these divergences cancel out in their difference, resulting in a finite result

$$\Delta\Gamma_b^{(1)} = \Gamma_{2b}^{(1)} - \Gamma_{1b}^{(1)} = -\frac{1.97J}{S}, \quad (\text{A-33})$$

Adding  $\Delta\Gamma_a^{(1)}$ , Eq.(A-21), to this result we obtain the total quantum correction  $\Delta\Gamma^{(1)} = \Delta\Gamma_a^{(1)} + \Delta\Gamma_b^{(1)} = -1.595J/S \approx -1.6J/S$ , as quoted in Eq.(10) of the main text.

#### Quantum corrections to $\Gamma_3$

Correction to  $\Gamma_3$  is in order of  $(1/S)^2$ , and to get such term in the ground state energy density we need to include both four-boson and six-boson terms in the Hamiltonian, Eqs. (A-2) and (A-3). We have

$$\begin{aligned} \mathcal{H}_i^{(4)} &= \frac{1}{8} \sum_{\mathbf{k}} (5J_{\mathbf{k}} - 2J_{\mathbf{Q}}) \left[ (\bar{\psi}_1^2 a_{\mathbf{Q}+\mathbf{k}} a_{\mathbf{Q}-\mathbf{k}} + \bar{\psi}_2^2 a_{-\mathbf{Q}+\mathbf{k}} a_{-\mathbf{Q}-\mathbf{k}}) + \text{h.c.} \right] \\ &\quad - \frac{1}{4} \sum_{\mathbf{k}} (J_{\mathbf{k}} - J_{\mathbf{Q}}) \left[ (\psi_0 \psi_2 a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger + \psi_0 \psi_1 a_{-\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger) + \text{h.c.} \right], \end{aligned} \quad (\text{A-34})$$

$$\mathcal{H}_i^{(6)} = \frac{1}{16S} \sum_{\mathbf{k}} \left( \frac{5}{2} J_{\mathbf{k}} - 4J_{\mathbf{Q}} \right) \left[ (\bar{\psi}_1 \psi_2^3 a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger + \bar{\psi}_1^3 \bar{\psi}_2 a_{-\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger) + \text{h.c.} \right]. \quad (\text{A-35})$$

We use the expression of  $\psi_0$  in Eq.(A-13), to rewrite  $\mathcal{H}_i^{(4)}$  as,

$$\begin{aligned} \mathcal{H}_i^{(4)} &= \frac{1}{8} \sum_{\mathbf{k}} (5J_{\mathbf{k}} - 2J_{\mathbf{Q}}) \left[ (\bar{\psi}_1^2 a_{\mathbf{Q}+\mathbf{k}} a_{\mathbf{Q}-\mathbf{k}} + \bar{\psi}_2^2 a_{-\mathbf{Q}+\mathbf{k}} a_{-\mathbf{Q}-\mathbf{k}}) + \text{h.c.} \right] \\ &\quad - \frac{1}{16S} \sum_{\mathbf{k}} (J_{\mathbf{k}} - J_{\mathbf{Q}}) \left[ (\bar{\psi}_1 \psi_2^3 a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger + \bar{\psi}_1^3 \bar{\psi}_2 a_{-\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger) + \text{h.c.} \right]. \end{aligned} \quad (\text{A-36})$$

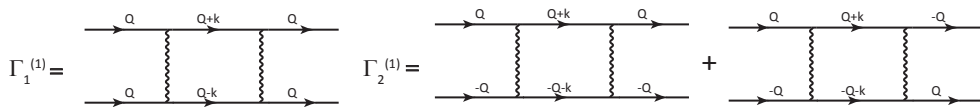


FIG. A-2. Diagrammatic representation of perturbative corrections to  $\Gamma_1$  and  $\Gamma_2$ .

The total perturbation Hamiltonian is now

$$\begin{aligned}\mathcal{H}_{i,k} &= \mathcal{H}_i^{(4)} + \mathcal{H}_i^{(6)} \\ &= \frac{1}{8} \sum_{\mathbf{k}} (5J_{\mathbf{k}} - 2J_{\mathbf{Q}}) \left[ (\bar{\psi}_1^2 a_{\mathbf{Q}+\mathbf{k}} a_{\mathbf{Q}-\mathbf{k}} + \bar{\psi}_2^2 a_{-\mathbf{Q}+\mathbf{k}} a_{-\mathbf{Q}-\mathbf{k}}) + \text{h.c.} \right] \\ &\quad - \frac{3}{32S} \sum_{\mathbf{k}} (J_{\mathbf{k}} - 2J_{\mathbf{Q}}) \left[ (\bar{\psi}_1 \psi_2^3 a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger + \psi_1^3 \bar{\psi}_2 a_{-\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger) + \text{h.c.} \right],\end{aligned}\quad (\text{A-37})$$

Because of two terms in (A-37), there are two contributions to  $\Delta E$  to order  $\psi^6/S^2$ . One comes from taking the product of  $\psi^2$  and  $\psi^4$  terms in the second-order perturbation theory. This yields

$$\begin{aligned}\Delta E_a &= -\frac{1}{2} \sum_{\mathbf{k},\mathbf{q}} \langle \mathcal{H}_{i,\mathbf{k}} \cdot \mathcal{H}_{i,\mathbf{q}} \rangle_0 = -\frac{3}{128S} \sum_{\mathbf{k},\mathbf{q}} (5J_{\mathbf{k}} - 2J_{\mathbf{Q}}) (J_{\mathbf{q}} - 2J_{\mathbf{Q}}) \times \\ &\quad \times \left[ \psi_1^3 \bar{\psi}_2^3 \langle a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger a_{\mathbf{Q}+\mathbf{q}} a_{\mathbf{Q}-\mathbf{q}} \rangle_0 + \bar{\psi}_1^3 \psi_2^3 \langle a_{-\mathbf{Q}+\mathbf{k}}^\dagger a_{-\mathbf{Q}-\mathbf{k}}^\dagger a_{-\mathbf{Q}+\mathbf{q}} a_{-\mathbf{Q}-\mathbf{q}} \rangle_0 \right]\end{aligned}\quad (\text{A-38})$$

and

$$\Delta \Gamma_{3,a}^{(2)} = -\frac{3}{64S^2} \sum_{\mathbf{k}} \frac{(5J_{\mathbf{k}} - 2J_{\mathbf{Q}})(J_{\mathbf{k}} - 2J_{\mathbf{Q}})}{J_0 - J_{\mathbf{k}}}.\quad (\text{A-39})$$

Diagrammatically, this correction to  $\Gamma_3$  is given by the first two diagrams in Fig.A-3,

Another contribution to  $\Delta E$  of order  $\psi^6/S^3$  comes from taking  $\psi^2$  term in (A-37) to 3rd order in perturbation theory. The corresponding term in the perturbative Hamiltonian (A-37) comes from fourth-order term in Holstein-Primakoff bosons and we write it separately:

$$\begin{aligned}\mathcal{H}_i^{(4)} &= \sum_{\mathbf{k}} \left[ \frac{1}{8} (5J_{\mathbf{k}} - 2J_{\mathbf{Q}}) (\psi_1^2 a_{\mathbf{Q}+\mathbf{k}}^\dagger a_{\mathbf{Q}-\mathbf{k}}^\dagger + \bar{\psi}_2^2 a_{-\mathbf{Q}+\mathbf{k}} a_{-\mathbf{Q}-\mathbf{k}}) + \text{h.c.} \right] \\ &\quad + \sum_{\mathbf{k}} \frac{3}{2} J_{\mathbf{Q}-\mathbf{k}} (\psi_1 \bar{\psi}_2 a_{\mathbf{k}}^\dagger a_{\mathbf{Q}+\mathbf{k}} + \text{h.c.}).\end{aligned}\quad (\text{A-40})$$

The third-order perturbative correction to the ground state density is

$$\begin{aligned}\Delta E_b &= \frac{1}{3!} \sum_{\mathbf{k},\mathbf{q},\mathbf{l}} \langle \mathcal{H}_{i,\mathbf{k}} \cdot \mathcal{H}_{i,\mathbf{q}} \cdot \mathcal{H}_{i,\mathbf{l}} \rangle_0 \\ &= \frac{3}{128} \sum_{\mathbf{k},\mathbf{q},\mathbf{l}} \frac{3}{2} J_{\mathbf{Q}-\mathbf{k}} (5J_{\mathbf{q}} - 2J_{\mathbf{Q}}) (5J_{\mathbf{l}} - 2J_{\mathbf{Q}}) (\psi_1^3 \bar{\psi}_2^3 + \text{h.c.}) \langle a_{\mathbf{k}}^\dagger a_{\mathbf{Q}+\mathbf{q}}^\dagger a_{\mathbf{Q}-\mathbf{q}}^\dagger a_{\mathbf{Q}+\mathbf{k}} a_{-\mathbf{Q}+\mathbf{l}} a_{-\mathbf{Q}-\mathbf{l}} \rangle_0\end{aligned}\quad (\text{A-41})$$

This leads to second  $1/S^2$  contribution to  $\Gamma_3$  in the form

$$\Gamma_{3b}^{(2)} = \frac{3}{32S^2} \sum_{\mathbf{k}} \frac{J_{\mathbf{Q}-\mathbf{k}} (5J_{\mathbf{k}} + J_0) (5J_{\mathbf{Q}+\mathbf{k}} + J_0)}{(J_0 - J_{\mathbf{k}}) (J_0 - J_{\mathbf{Q}+\mathbf{k}})}.\quad (\text{A-42})$$

In diagrammatic approach, this correction comes from the third diagram in Fig.A-3,

The total  $\Gamma_3^{(2)}$  is the sum of terms in Eqs.(A-39) and Eq.(A-42)

$$\Gamma_3^{(2)} = \frac{3}{32S^2} \sum_{\mathbf{k}} \left( \frac{J_{\mathbf{Q}-\mathbf{k}} (5J_{\mathbf{k}} + J_0) (5J_{\mathbf{Q}+\mathbf{k}} + J_0)}{(J_0 - J_{\mathbf{k}}) (J_0 - J_{\mathbf{Q}+\mathbf{k}})} - \frac{(5J_{\mathbf{k}} + J_0) (J_{\mathbf{k}} + J_0)}{2(J_0 - J_{\mathbf{k}})} \right) = -\frac{0.97J}{S^2}.\quad (\text{A-43})$$

Here again we observe the cancellation of logarithmic singularities, present in the individual integrals.

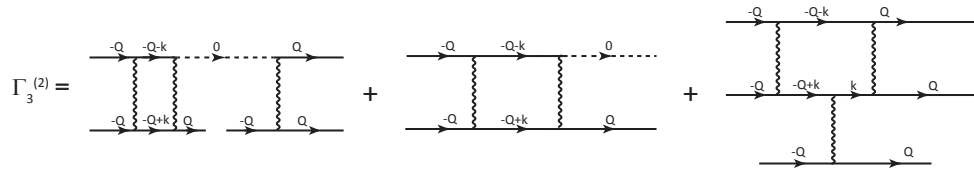


FIG. A-3. Diagrams for  $1/S$  corrections to  $\Gamma_3$ . The first two diagrams are 2nd order perturbation corrections from the product of  $\psi^2$  and  $\psi^4$  terms in Eq (A-37), the last diagram is 3th order perturbative correction from (A-40).

### INTERMEDIATE DOUBLE CONE STATE FOR $J - J'$ MODEL

In this Section, we analyze the phase transition from the cone to the coplanar state, when magnetic field  $h$  is below  $h_{\text{sat}}$ , i.e.,  $\mu = h_{\text{sat}} - h$  is positive. We remind that at  $\mu = 0+$ , the cone state is stable at  $\delta J = J - J' > \delta J_c = 0.42J/\sqrt{S}$ . Accordingly, we treat  $\delta J \approx \delta J_c$  as a small parameter.

Our goal will be to obtain the spin-wave spectrum in the cone state to leading order in  $\delta J$  and with quantum corrections. The magnon modes in the cone state are

$$a_{\mathbf{k}} = \sqrt{N}\psi_1\delta_{\mathbf{k},\mathbf{Q}} + \tilde{a}_{\mathbf{k}}. \quad (\text{A-44})$$

where, we remind,  $\tilde{a}_{\mathbf{k}}$  describe non-condensed bosons and  $\psi_1 \propto \sqrt{S}$  describes the condensate fraction.

We first consider classical spin-wave excitations at the leading order in  $1/S$ , but a non-zero  $\delta J$ , and then add quantum  $1/S$  corrections to the excitation spectrum. As before, the latter already contain  $1/S$  and can be computed in the isotropic  $\delta J = 0$  limit.

#### Classical spin-wave excitations

Spatially anisotropic Hamiltonian to second order in  $\tilde{a}_{\mathbf{k}}$  reads

$$\begin{aligned} \mathcal{H}_{\text{anis}} &= H_1 + H_2 \\ H_1 &= \mathcal{H}_{\text{anis}}^{(2)} = \sum_{\mathbf{k}} \left[ S(\tilde{J}_{\mathbf{k}} - \tilde{J}_{\mathbf{Q}}) - \mu \right] \tilde{a}_{\mathbf{k}}^\dagger \tilde{a}_{\mathbf{k}}, \end{aligned} \quad (\text{A-45})$$

$$\begin{aligned} H_2 &= \frac{1}{8} \sum_{\mathbf{q}} \left[ (5\tilde{J}_{\mathbf{q}} - 2\tilde{J}_{\mathbf{Q}}) \psi_1^2 \tilde{a}_{\mathbf{Q}+\mathbf{q}}^\dagger \tilde{a}_{\mathbf{Q}-\mathbf{q}} + \text{h.c.} \right] \\ &\quad + \sum_{\mathbf{k}} (\tilde{J}_0 - \tilde{J}_{\mathbf{Q}} + \tilde{J}_{\mathbf{Q}-\mathbf{k}} - \tilde{J}_{\mathbf{k}}) |\psi_1|^2 \tilde{a}_{\mathbf{k}}^\dagger \tilde{a}_{\mathbf{k}}, \end{aligned} \quad (\text{A-46})$$

where, we remind,  $\tilde{J}_{\mathbf{k}}$ , where  $\tilde{J}_{\mathbf{k}} = 2(J \cos[k_x] + 2J' \cos[\frac{k_x}{2}] \cos[\frac{\sqrt{3}k_y}{2}])$ .  $\tilde{J}_{\mathbf{k}}$  has minimum  $\tilde{J}_{\mathbf{Q}}$  at  $\mathbf{Q} = (Q_i, 0)$ , and  $Q_i = 2 \cos^{-1}[-J'/2J]$ . At small  $\delta J \sim \delta J_c$ ,  $\mathbf{Q}$  by  $\mathbf{Q} \approx (4\pi/3 - \Delta Q, 0)$ , where  $\Delta Q = 4\pi/3 - Q_i = 2\delta J/\sqrt{3}$ .

Our goal is to obtain the renormalization of the excitation spectrum  $\omega_k$  to second order in the condensate, i.e., to order  $\psi^2$ . The first term in  $H_2$  is irrelevant for this purpose as it describes excitations with momentum transfer  $2\mathbf{Q}$ , which can only contribute to  $\omega_k$  at second order in perturbation theory, but such term will be of order  $\psi^4$ . The remaining term in  $H_2$  is quadratic in non-condensed bosons and directly contribute to spin-wave spectrum to second order in  $\psi$ .

We will be interested in magnon excitations for  $\mathbf{k}$  near  $-\mathbf{Q} = -(Q_i, 0)$ . Accordingly, we set  $\mathbf{k} = -\mathbf{Q} + \mathbf{p}$  and treat  $\mathbf{p}$  as small momentum. Restricting with small  $\mathbf{p}$  and using the approximate form of  $\mathbf{Q}$ , we re-write Eqs.(A-45) and (A-46) as

$$\mathcal{H}_{\text{anis}} = \sum_{\mathbf{p}} \left[ \frac{3}{4} S J (p_x^2 + p_y^2) + J |\psi_1|^2 \left( \frac{h_{\text{sat}}}{S J} + \frac{9}{2} p_x \Delta Q + \frac{27}{4} (\Delta Q)^2 \right) - \mu \right] \tilde{a}_{-\mathbf{Q}+\mathbf{p}}^\dagger \tilde{a}_{-\mathbf{Q}+\mathbf{p}}, \quad (\text{A-47})$$

where  $h_{\text{sat}} = S(\tilde{J}_0 - \tilde{J}_{\mathbf{Q}}) = S\Gamma_1^{(0)}$ . Completing the square and rearranging, and setting  $\mathbf{k} = -\mathbf{Q} + \mathbf{p}$  again, we obtain

$$\mathcal{H}_{\text{anis}} = \sum_{\mathbf{k}} S \omega_{\mathbf{k}}^{(1)} \tilde{a}_{\mathbf{k}}^\dagger \tilde{a}_{\mathbf{k}}, \quad (\text{A-48})$$

where

$$\omega_{\mathbf{k}}^{(1)} = \frac{3}{4} J \left[ (k_x + \bar{Q}_i)^2 + k_y^2 + \varepsilon_{\text{min}} \right], \quad (\text{A-49})$$

$$\varepsilon_{\text{min}} = 9 \frac{|\psi_1|^2}{S} \left( 1 - \frac{|\psi_1|^2}{S} \right) (\Delta Q)^2 + \frac{4}{3} \frac{1}{S J} \left( \frac{|\psi_1|^2}{S} h_{\text{sat}} - \mu \right). \quad (\text{A-50})$$

Here  $\bar{Q}_i = 4\pi/3 - \Delta Q + 3|\psi_1|^2 \Delta Q/S$ , and the minimum of  $\omega_{\mathbf{k}}^{(1)}$  is at  $(-\bar{Q}_i, 0)$ .

In the classical approximation (leading order in  $1/S$ ), the condensate density is  $|\psi_1|^2/S = \mu/(S\Gamma_1^{(0)}) = \mu/h_{\text{sat}}$ , and we obtain

$$\varepsilon_{\text{min,class}} = \frac{12\mu}{h_{\text{sat}} J^2} \frac{h}{h_{\text{sat}}} (\delta J)^2. \quad (\text{A-51})$$

To this order, the second term in (A-50) nullifies exactly. To the same accuracy,  $\bar{Q}_i = Q_i + (4\pi/3 - Q_i)(3\mu/h_{\text{sat}}) + O(1/S)$ .

### Quantum corrections

Since at  $\mu = 0$  the critical value of  $\delta J_c \sim 1/\sqrt{S}$ , we recognize that in fact  $\varepsilon_{\min, \text{class}} \sim 1/S$  in the relevant range of  $\delta J$ , where the transition between the cone and the coplanar state takes place. This means that Eq.(A-51) is not complete – one needs to add to it quantum  $1/S$  contributions. These come from several sources as we now describe.

The first quantum correction comes from the fact that the relation between the condensate wave function  $\psi_1$  and  $\Gamma_1$ :

$$\frac{|\psi_1|^2}{S} = \frac{\mu}{S\Gamma_1} \quad (\text{A-52})$$

contains  $1/S$  terms because  $\Gamma_1 = \Gamma_1^{(0)} + \Gamma_1^{(1)}$ , where  $\Gamma_1^{(0)} = h_{\text{sat}}/S = \tilde{J}_0 - \tilde{J}_{\mathbf{Q}} \sim J$  represents classical ( $S = \infty$ ) contribution already accounted for in deriving (A-51), while  $\Gamma_1^{(1)} = \Gamma_{1a}^{(1)} + \Gamma_{1b}^{(1)} \sim J/S$  represents the leading  $1/S$  correction to it. The term with subindex  $a$  describes contribution from normal ordering,  $-J_{\mathbf{Q}}/(8S)$  in (A-21), while the one with subindex  $b$  describes the contribution from quantum fluctuations, Eq.(A-32).

Hence, in the cone state,

$$\frac{|\psi_1|^2}{S} = \frac{\mu}{S\Gamma_1} = \frac{\mu}{S(\Gamma_1^{(0)} + \Gamma_1^{(1)})} = \frac{\mu}{h_{\text{sat}}} \left(1 - \frac{\Gamma_1^{(1)}}{\Gamma_1^{(0)}}\right) \quad (\text{A-53})$$

contains quantum correction,  $\sim \Gamma_1^{(1)}$ . Substituting the full form of  $\psi$  into Eq. (A-50) and collecting  $1/S$  terms we obtain first  $1/S$  correction  $\Delta\varepsilon_{\min,1}$

$$\Delta\varepsilon_{\min,1} = -\frac{4}{3} \frac{\mu}{h_{\text{sat}}J} \Gamma_1^{(1)} + O(1/S^2), \quad (\text{A-54})$$

The two other quantum corrections are associated with  $\Gamma_2$  processes. One  $\Gamma_2$  correction comes from Eq.(A-18), which, we remind, emerges when we normal order bosonic operators in the Holstein-Primakoff transformation. It is easiest to obtain this contribution via a real-space representation

$$a_{\mathbf{r}} = \psi_1 e^{i\mathbf{Q}\cdot\mathbf{r}} + \tilde{a}_{\mathbf{r}}, \quad (\text{A-55})$$

where, as before,  $\tilde{a}_{\mathbf{r}}$  describes non-condensate magnons. Substituting this into (A-18) we obtain

$$\delta\mathcal{H}^{(4)} = -\frac{|\psi_1|^2}{8S} \sum_{\mathbf{k}} (\tilde{J}_{\mathbf{k}} + \tilde{J}_{\mathbf{Q}}) \tilde{a}_{\mathbf{k}}^\dagger \tilde{a}_{\mathbf{k}} \approx -\frac{|\psi_1|^2}{4S} \sum_{\mathbf{p}} \tilde{J}_{\mathbf{Q}} \tilde{a}_{-\mathbf{Q}+\mathbf{p}}^\dagger \tilde{a}_{-\mathbf{Q}+\mathbf{p}}, \quad (\text{A-56})$$

for  $\mathbf{k} \approx -\mathbf{Q}$ . Adding this to (A-50) we obtain a  $\Gamma_{2a}^{(1)}$  correction to  $\varepsilon_{\min}$ ,

$$\Delta\varepsilon_{\min,2} = \frac{4}{3J} \frac{\mu}{h_{\text{sat}}J} \left(-\frac{\tilde{J}_{\mathbf{Q}}}{4S}\right) = \frac{4}{3} \frac{\mu}{h_{\text{sat}}J} \Gamma_{2a}^{(1)}. \quad (\text{A-57})$$

Observe that, because we already have  $1/S$  in the prefactor, we can neglect the difference between  $\tilde{J}_{\mathbf{Q}}$  and  $J_{\mathbf{Q}}$ .

The third quantum correction (also associated with  $\Gamma_2$ ) comes from terms cubic in non-condensate magnons  $\tilde{a}_{\mathbf{k}}$  taken to second order in perturbation theory. The cubic terms are generated from (A-2) via the substitution (A-44). Such terms are necessarily linear in  $\psi_1$ :

$$H_3 = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}, \mathbf{q}} V_{\mathbf{q}}(\mathbf{k}, \mathbf{Q}) \left( \psi_1 \tilde{a}_{\mathbf{Q}-\mathbf{q}}^\dagger \tilde{a}_{\mathbf{k}+\mathbf{q}}^\dagger \tilde{a}_{\mathbf{k}} + \text{h.c.} \right). \quad (\text{A-58})$$

A second-order in perturbation theory in (A-58) produces a  $1/S$  correction to the dispersion of  $\tilde{a}_{\mathbf{k}}$  magnons with  $\mathbf{k} \approx -\mathbf{Q}$  in the form

$$\begin{aligned} \Delta\varepsilon_{\min,3} &= -\frac{4}{3} \frac{1}{JS} \sum_{\mathbf{q}, \mathbf{q}'} V_{\mathbf{q}}(-\mathbf{Q}, \mathbf{Q}) V_{\mathbf{q}'}(-\mathbf{Q}, \mathbf{Q}) |\psi_1|^2 \langle \tilde{a}_{\mathbf{Q}-\mathbf{q}}^\dagger \tilde{a}_{-\mathbf{Q}+\mathbf{q}}^\dagger \tilde{a}_{\mathbf{Q}-\mathbf{q}'} \tilde{a}_{-\mathbf{Q}+\mathbf{q}'} \rangle_0, \\ &= \frac{4}{3} \frac{|\psi_1|^2}{SJ} \Gamma_{2b}^{(1)} \approx \frac{4}{3} \frac{\mu}{h_{\text{sat}}J} \Gamma_{2b}^{(1)}. \end{aligned} \quad (\text{A-59})$$

Adding Eqs. (A-54), (A-57), and (A-59) to the classical result for  $\varepsilon_{\min}$ , we obtain the final expression for the minimal energy  $\varepsilon_{\min}$  of the magnons at  $\mathbf{k} \approx -\mathbf{Q}$ :

$$\varepsilon_{\min, \text{tot}} = \frac{12\mu}{h_{\text{sat}} J^2} \left[ \frac{h}{h_{\text{sat}}} (\delta J)^2 + \frac{\Gamma_2^{(1)} - \Gamma_1^{(1)}}{9} \right] = \frac{12\mu}{h_{\text{sat}} J^2} \left[ \frac{h}{h_{\text{sat}}} (\delta J)^2 - (\delta J_c)^2 \right] \approx \frac{12\mu}{h_{\text{sat}} J^2} \left[ (\delta J)^2 - (\delta J_c)^2 \left(1 + \frac{\mu}{h_{\text{sat}}}\right) \right]. \quad (\text{A-60})$$

Observe that  $\Gamma_2^{(1)} - \Gamma_1^{(1)} = \Delta\Gamma^{(1)} = -1.6J/S$  and  $\delta J_c = \sqrt{1.6J^2/(9S)} \approx 0.42J/\sqrt{S}$ , see Eq.(10) and description below it in the main text.

At  $\mu = +0$ , magnon energy of  $\mathbf{k} = -\mathbf{Q}_i$  vanishes at  $\delta J = \delta J_c$ , as expected. However, at a finite  $\mu$ , the instability occurs at  $\delta J_h = h_{\text{sat}} \delta J_c / h > \delta J_c$  and the mode that condenses carries momentum  $\mathbf{k} = (-\bar{Q}_i, 0) \neq -\mathbf{Q}_i$  different from  $-\mathbf{Q}_i$ . This gives rise to the development of the second condensate with momentum  $(-\bar{Q}_i, 0)$ . The resulting state is the double cone phase described in the main text.

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